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LIGHT-DIFFUSING SYNTHETIC RESINS  
[Kokakusansei goseijjushi]

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## Claims

1. Light-diffusing synthetic resins are characterized in that silicone resin fine particles having average particle diameter of 1-6  $\mu\text{m}$  are dispersed in transparent synthetic resins.

2. Light-diffusing synthetic resins are characterized in that silicone resin fine particles having average particle diameter of 1-6  $\mu\text{m}$  and inorganic transparent powder having particle diameter of 1-7  $\mu\text{m}$  are dispersed in transparent synthetic resins.

3. Light-diffusing synthetic resins described in Claim 1 or 2, wherein the transparent synthetic resins are methacrylic resins.

## Detailed explanation of invention

### Industrial application field

The present invention relates to light-diffusing resins, more specifically to light-diffusing synthetic resins suitable for illumination covers, illumination signboards, displays, glazing and the like.

### Prior art

As characteristics desired in light-diffusing synthetic resins useful as molding materials of lighting apparatus, various displays and the like, high light diffusivity and high light transmittance are desired even from the point of effective use of electric energy, and examples of such light-diffusing synthetic resins include (a) transparent resins containing inorganic powders such as barium sulfate, calcium carbonate and the like with average particle diameter of 10  $\mu\text{m}$  or less (Japanese Kokai Patent Sho 60[1985]-139758), (b) those obtained by dispersing inorganic transparent powders, such as glass powder, quartz powder, calcium fluoride of 1-10  $\mu\text{m}$  and the like, or organic transparent powders, such as polystyrene, polymethacrylate, and acrylic ester fluoride of 1-10  $\mu\text{m}$ , in transparent resins (Japanese

Kokoku Patent Sho 60[1985]-21662), (c) those obtained by dispersing transparent fine particles, such as silica, glass, calcium fluoride, aluminum hydroxide of 4-50  $\mu\text{m}$  and the like, in transparent resins (Japanese Kokai Patent Sho 60[1985]-139758, Japanese Kokai Patent Sho 60[1985]-184559 and Japanese Kokai Patent Sho 61[1986]-4762) and the like.

#### Problem to be solved by invention

In the aforementioned conventional light-diffusing synthetic resins, however, if the additional amount of the powders is reduced in order to enhance the total light transmittance, the haze (haze value) is lowered and the lighting of the inner part becomes transparent since inorganic powders as light-diffusing powder are mainly added. On the other hand, if the additional amount is increased to raise the haze for preventing the formation of transparency, the total light transmittance is lowered and abrasion and damage of molding machines, nozzles, dies, molds and the like easily occur since hard inorganic powders are used. Particularly in the case of plate making by extrusion molding process, drawbacks such as die lines easily form when the particle diameter of powders is large or small so that there arises such a problem of difficulty in continuous operation for long time. Thus, those can not sufficiently meet the market need for arrival of light-diffusing resins having high light diffusivity and high light transmittance.

Therefore, the objective of the present invention is to provide light-diffusing resins having high light diffusivity and high light transmittance and also suitable for practical productivity.

#### Means to solve the problem

Present inventor et al., had assiduously studied various light diffusion agents to solve the aforementioned problem and, as a result, it was found that high light diffusivity and high light

transmittance are attained by including silicone resin particles having specified particle diameter in transparent synthetic resins. Thereby, the present invention was completed.

Namely, the gist of the present invention is light-diffusing synthetic resins obtained by dispersing silicone resin particles having average particle diameter of 1-6  $\mu\text{m}$  in transparent synthetic resins and also light-diffusing synthetic resins obtained by dispersing silicone resin particles having average particle diameter of 1-6  $\mu\text{m}$  and inorganic transparent material powders having average particle diameter of 1-7  $\mu\text{m}$  in transparent synthetic resins.

Hereinafter the present invention is explained in detail.

Transparent synthetic resins to be used in the present invention are not restricted specifically and, for instance, methacrylic resins, styrene resins, polycarbonate resins and the like are exemplified, particularly methacrylic resins containing methyl methacrylate as the main component and having high light transmittance are preferably used.

Silicone resin fine particles to be used in the present invention are fine particles of silicone resins such as polymethyl silsesquioxane comprising three functional units and prepared by Building-up process, and those are solvent-insoluble, heat-infusible polymer particles hardened by forming dense siloxane bonds. Those particles have average particle diameter (weight-average particle diameter) of 1-6  $\mu\text{m}$ , preferably 2-4  $\mu\text{m}$  and as their shape, truly spherical rather than amorphous is preferred since higher haze is obtained and transparency formation is difficult. When the average particle diameter of the fine particles is smaller than 1  $\mu\text{m}$ , transmitted light becomes reddish since selection of transmission wavelength occurs during transmitting of light, and this is not preferred. On the other hand, when it exceeds 6  $\mu\text{m}$ , transparency easily forms and it is seen such a tendency makes it difficult to achieve both properties of high light diffusivity and high light transmittance.

Since light-diffusing resins should have haze of at least about 90%, preferably at least 92%, from aspect of practical performance such as not seeing the lighting of the inner part, namely not being transparent, in conventional resin system containing barium sulfate (3  $\mu\text{m}$ ), calcium carbonate (5  $\mu\text{m}$ ) and glass beads (3  $\mu\text{m}$ ), total light transmittance of about 65-75% at the most is obtained unless the surface is made matt to cause surface scattering.

In contrast to this, since light-diffusing synthetic resins of the present invention are resins obtained by dispersing silicone resin fine particles having particle diameter of 1-6  $\mu\text{m}$ , they have haze of about 94% even when total light transmittance is about 86%, and light-diffusing synthetic resins having beautiful gloss showing high light diffusivity and high light transmittance can be obtained without being transparent.

It is reasonable that the additional amount of the aforementioned silicone resin fine particles to be used in the present invention is limited to about 5 parts by weight per 100 parts by weight of the resins when the following matters are considered: it should not cause surface roughening of molded articles; the impact strength should not be lowered to such a level that it is practically inconvenient; secondary processing such as vacuum molding and the like can be easily carried out; and economy. When the plate thickness is considered, the amount of 6-40  $\text{g/m}^2$ , preferably 8-30  $\text{g/m}^2$ , is desired.

Furthermore, inorganic transparent powders to be used in the present invention are contained in order to enhance whiteness of light-diffusing synthetic resins particularly at reflected light, and barium sulfate, calcium carbonate and the like are preferably used. The particle diameter of these powders is 1-7  $\mu\text{m}$ , preferably 2-5  $\mu\text{m}$ , and the additional amount of the powders is, per 100 parts by weight of resins, 2 parts by weight or less, preferably 0.3-1 part by weight. It is desired that the additional amount be less than the additional amount of the aforementioned silicone resin fine particles.

As mentioned above, light-diffusing synthetic resins of the present invention are suitable as molding materials of illumination covers, illumination signboards, displays, glazing and the like, and the aforementioned lighting apparatus, various displays and glazing articles can be produced by injection molding or extrusion.

#### Application examples

Hereinafter, the present invention is explained in detail by application examples.

In the application examples, "total light transmittance" and "haze" were measured by integrating sphere type light transmittance measuring apparatus (RM-15, manufactured by Murakami Color Technical Research Laboratory) in accordance with JIS K6717.

"Brightness" is a, b system brightness measured by color difference meter (SM-2, manufactured by Suga Shikenki K.K.). Further, transparency, which was visual judgment of light diffusivity, was confirmed as follows: a sample piece is put in parallel 8 cm in front of a 10 W straight-tube fluorescent lamp, and the general view of the fluorescent lamp through a sample plate is visually checked at the position 1 m from the aforementioned sample piece. Its state is expressed by the following symbols.

- O The outline of the fluorescent lamp is not seen.
- Δ The outline of the fluorescent lamp is vaguely seen.
- x The outline of the fluorescent lamp is clearly seen.

Furthermore, particle diameter in the application examples means weight-average particle diameter, and it is measured by particle size distribution measuring apparatus (SKA-5000, manufactured by Seishin Enterprise K.K.).

### Application Examples 1-3

Methyl acrylate-methyl methacrylate copolymer with methyl acrylate content of 5% and molecular weight of 160,000 as methacrylic resin 100 parts by weight was mixed with truly spherical silicone resin having particle diameter of 2.3  $\mu\text{m}$  at various additional amounts, stirred, extruded by kneading-melt extrusion die at a velocity of 10 kg/H in a single-screw extruder to sheets, polished by poly-sink roll and cooled to obtain flat plate-form molded articles having thickness of 1-3 mm and having good appearance. Non-transparent light-diffusing resin plates having total light transmittance 86% and haze 94% were obtained, and the performance evaluation results of those molded articles are shown in Table 1.

### Application Examples 4-6

Flat plate-form molded articles having good appearance were obtained by the same manner as in Application Examples 1-3 except that truly spherical silicone resin fine particles having particle diameter 3.6  $\mu\text{m}$  (Tospearl 130, manufactured by Toshiba Silicone Co., Ltd.) were used. The performance evaluation results of those molded articles are shown in Table 1. It is seen from these results that non-transparent light-diffusing resin plates having total light transmittance 86% were obtained.

### Application Examples 7, 8

Flat plate-form molded articles having good appearance were obtained by the same manner as in Application Examples 1-3 except that amorphous silicone resin fine particles having particle diameter 5.9  $\mu\text{m}$  were used. The performance evaluation results of those molded articles are shown in Table 1. It is seen from the results that the results are not better than those obtained by using truly spherical silicone



resin fine particles, but there is remarkable improvement as compared with conventional diffusive resin plates.

#### Application Examples 9-11

Flat plate-form molded articles having good appearance were obtained by the same manner as in Application Examples 1-3 except that a mixture of truly spherical silicone resin having particle diameter of 3.6  $\mu\text{m}$  and barium sulfate having particle diameter of 3  $\mu\text{m}$  and a mixture of amorphous silicone resin having particle diameter of 5.9 [ $\mu\text{m}$ ] and calcium carbonate having particle diameter of 5  $\mu\text{m}$  were used. The performance evaluation results of those molded articles are shown in Table 1. It is seen from the results that the brightness is improved without lowering much the total light transmittance.

#### Comparative Examples 1-3

Flat plate-form molded articles were obtained by the same manner as in Application Examples 1-3 except that precipitated barium sulfate having particle diameter of 3  $\mu\text{m}$  was used. The performance evaluation results of those molded articles are shown in Table 1. It is seen from the results that appearance drawbacks, called die lines, tend to appear as the additional amount of barium sulfate increases.

#### Comparative Examples 4-6

Flat plate-form molded articles were obtained by the same manner as in Application Examples 1-3 except that calcium carbonate having particle diameter of 5  $\mu\text{m}$  was used. The performance evaluation results of those molded articles are shown in Table 1. It is seen from the results that die lines tend to easily appear even when the additional amount is little.

TABLE 1

(1)	(3) 微粒子の種類 (粒子径)	(13) 微粒子の添加量 (g/ml)	(14) 膜厚 (nm)	(15) 光拡散性	(16) 全光線透過率 (%)	(17) ヘイズ値 (%)	(18) 白・色度
実施例 1	シリコーン樹脂 (2.3 $\mu\text{m}$ )	(4) 9	1	○	86	94	
2		18	2	○	81	95	
3		27	3	○	73	95	
4	シリコーン樹脂 (3.6 $\mu\text{m}$ )	(5) 9	1	○	86	93	19
5		18	2	○	84	95	23
6		27	3	○	77	95	24
7	シリコーン樹脂 (5.9 $\mu\text{m}$ )	(6) 18	2	○	78	94	26
8		27	3	○	71	95	27
9	シリコーン樹脂 (3.6 $\mu\text{m}$ )	(7) 9	1	○	82	93	30
10	硫酸バリウム (3 $\mu\text{m}$ )	(8) 5.4					
		9	1	○	80	94	33
11	シリコーン樹脂 (5.9 $\mu\text{m}$ )	(9) 9.1	1	○	81	93	30
	炭酸カルシウム (5 $\mu\text{m}$ )	(10) 5.4					
比較例 1	硫酸バリウム (3 $\mu\text{m}$ )	(11) 16	1	○	73	94	41
(2) 2		32	2	○	62	95	47
3		49	3	○	54	96	48
4	炭酸カルシウム (5 $\mu\text{m}$ )	(12) 8	1	×	79	82	22
5		16	3	○	70	91	26
6		24	3	○	64	94	35

- Key: 1 Application Example \_\_\_\_
- 2 Comparative Example \_\_\_\_
- 3 Type of fine particles (particle diameter)
- 4 Silicone resin (2.3  $\mu\text{m}$ )
- 5 Silicone resin (3.6  $\mu\text{m}$ )
- 6 Silicone resin (5.9  $\mu\text{m}$ )
- 7 Silicone resin (3.6  $\mu\text{m}$ )
- 8 Barium sulfate (3  $\mu\text{m}$ )

- 9      Silicone resin (5.9  $\mu\text{m}$ )
- 10     Calcium carbonate (5  $\mu\text{m}$ )
- 11     Barium sulfate (3  $\mu\text{m}$ )
- 12     Calcium carbonate (5  $\mu\text{m}$ )
- 13     Additional amount of fine particles ( $\text{g/m}^3$ )
- 14     Plate thickness (mm)
- 15     Light diffusivity
- 16     Total light transmittance (%)
- 17     Haze (%)
- 18     Brightness

#### Effect of the invention

As stated above, since light-diffusing synthetic resins of the present invention are synthetic resins obtained by dispersing silicone resins having average particle diameter of 1-6  $\mu\text{m}$  in transparent resins, it is not necessary to use hard inorganic powders, and there is no concern of abrasion and damage of molding machines, dies and the like. Furthermore, since the particle diameter is small, it is hard to form appearance drawback such as die lines; further, those are superior in actual productivity and also possess high light diffusivity and high light transmittance so that those are useful as light-diffusing synthetic resins and suitable as molding materials of illumination covers, illumination signboards, displays, glazing and the like for supplying their molded articles.

Furthermore, since light-diffusing synthetic resins of the present invention are synthetic resins obtained by dispersing silicone resins having average particle diameter of 1-6  $\mu\text{m}$  and inorganic transparent materials having average particle diameter of 1-7  $\mu\text{m}$  in transparent resins, the brightness can

be controlled by the additional amount of inorganic transparent materials so that light-diffusing synthetic resins having whiteness in reflecting beam and possessing high light diffusivity and high light transmittance can be supplied.